

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

Applicants : Tetsuya Fukunaga  
Serial No. : 09/701,585  
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For : CATALYST AND PROCESS  
FOR REFORMING HYDROCARBON  
Art Unit & Examiner : 1754, MEDINA SANABRIA,  
MARIBEL

DECLARATION UNDER 37 C.F.R. 1.132

Honorable Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Sir:

I, the undersigned Tetsuya Fukunaga, c/o Idemitsu Kosan Co., Ltd.  
1280, Kamiizumi, Sodegaura-shi, Chiba 299-0293 Japan do hereby declare  
that:

1. I completed the master course of Tokyo University in March 1987,  
and entered Idemitsu Kosan Co., Ltd. in April 1987.

I work at catalyst and reaction design laboratory in Central  
Research Laboratories of Idemitsu Kosan Co., Ltd. and am specialized in  
chemical engineering.

2. I am an inventor of present U.S. Patent Application as identified  
above and familiar with the subject matter disclosed in the application.

3. Experiment

Object of the Experiment

In order to clarify an advantageous effect of the catalyst containing  
zirconium in an amount from 0.05-20 weight % as reduced to ZrO<sub>2</sub> by the  
present invention, the following Experiment was carried out.

Reference Experiment

## (1) Preparation of catalyst

The preparation of catalyst of Example 4 described in the present specification was carried out except that five kinds of aqueous solution changing concentration of zirconium oxychloride  $[\text{ZrO}(\text{OH})\text{Cl}]$  (Zr content as reduced to  $\text{ZrO}_2$  (0~12g)) were used, and that a carrier ( $\alpha$ -alumina) was impregnated with the above aqueous solution respectively, to thereby obtain catalysts IV-1 to IV-5. The composition of the catalysts are shown in Table A.

Table A Compositions of alumina

No.	1	2	3	4	5	6
Catalyst	IV-1	IV-2	IV**	IV-3	IV-4	IV-5
Metal component(wt.%)						
Zirconium*	0	2.0	5.0	10.0	17.0	24.0
Ruthenium	0.5	0.5	0.5	0.5	0.5	0.5
Cobalt	1.0	1.0	1.0	1.0	1.0	1.0

\*Zirconium: as reduced to  $\text{ZrO}_2$

\*\*the catalyst in Example 4 of the present specification (see Table 2)

## (2) Production of hydrogen

The same autothermal reforming reaction as in Example 4 of the present specification was carried out except that the catalysts were changed to above-obtained catalysts IV-1 to IV-5 respectively. The results of reaction by the catalyst are shown in Table B.

Table B Results of autothermal reforming

No.	Catalyst	HC conversion(%*)	Decomposition of carbon**
1	IV-1	79.8	0.7
2	IV-2	86.6	0.1
3	IV***	91.7	0.1
4	IV-3	90.2	0.1
5	IV-4	83.5	0.1
6	IV-5	78.8	0.1

\*HC conversion % =  $[1 - (\text{no. of carbon atoms of hydrocarbon in product}) / (\text{no. of carbon atoms of hydrocarbon in feedstock})]$

\*\*Relative amount of carbon deposited on catalyst after reaction for 10 hours (the amount in Comp.Ex.1 of the present specification is 1.0)

\*\*\* the catalyst in Example 4 of the present specification (see Table 6)

### (3) Reformation of hydrocarbon using carbon dioxide

The same Reformation of hydrocarbon as in Example 9 of the present specification was carried out except that the catalyst were changed to above-obtained catalysts IV-1 to IV-5 respectively. The results of reaction are shown in Table C.

Table C Results of reforming reaction of hydrocarbon

No.	Catalyst	CO yield (mol%)*	
		After 1 h	After 10 h
1	IV-1	75	67
2	IV-2	82	81
3	XIV**	85	83
4	IV-3	85	83
5	IV-4	81	79
6	IV-5	78	75

\*(amount of CO in product: mol)/(amount of CO<sub>2</sub> + CH<sub>4</sub> in feedstock: mol) × 100

\*\*the catalyst in Example 12 of the present specification (see Tables 8, 11)

### Consideration

From the above-results, it is clearly understandable that the catalyst comprising an inorganic oxide carrier carrying zirconium and ruthenium which contains zirconium in an amount from 0.05-20 wt. % as reduced to ZrO<sub>2</sub> by the present invention is superior to a catalyst containing zirconium outside the ranges.

Namely,

- ① High conversion is provided in autothermal reforming reaction of desulfurized naphtha and only a small amount of coke is deposited on the catalyst after autothermal reforming reaction for 10 hours.(see Table B)
- ② High yield of carbon monoxide is provided in hydrocarbon reforming reaction making use of carbon dioxide, and the yield remains constant.(see Table C)

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Tetsuya Fukunaga  
Tetsuya Fukunaga

Date: 1. 7. 2003